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Assessment of ozone and UV pre-oxidation processes for mitigating microbiologically accelerated monochloramine decay

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ABSTRACT

This paper reports the effects of pre-oxidation processes including ozone and ultraviolet (UV) irradiation prior to chloramination on microbiologically assisted monochloramine decay. Water samples with varying water qualities were pre-oxidated by ozone and UV irradiation, followed by determination of chemical and microbiological monochloramine decay (F_m). Both ozone and UV could effectively improve the reduction of microbial-like compounds responsible for monochloramine decay in treated water samples. Reductions in F_m values were observed for ozone contact times (Ct) greater than 5 mg min/L and UV doses greater than 30 mJ/cm^2. However, UV was less effective than ozone in reducing F_m values in raw water samples. Complete removal of the microbiological component of the decay for raw water samples was not found either with ozone (even with Ct of 50 mg min/L) or UV (even with UV dose of 120 mJ/cm^2). The effects of pre-oxidation processes on chemical (k_c) and microbial (k_m) decay coefficients were assessed. Increasing both ozone Ct and UV dose ahead of chloramine did not affect the chemical decay component, but they changed the microbiological component of decay. Changes in organic matter after ozonation were also characterized using three-dimensional fluorescence excitation–emission matrix (3D-FEEM) spectroscopy, and correlations between 3D-FEEM spectroscopy results and F_m values were found. Intensities of humic-like, fulvic-like, microbial protein-like and aromatic protein-like substances were reduced by pre-ozonation. Based on the 3D-FEEM results, we can confirm that the slowdown of monochloramine decay rate is due to the reduction of marine humic-like substances and soluble microbial protein-like compounds by pre-ozonation.

1. Introduction

Monochloramine is a commonly used disinfectant in drinking water treatment which is often used when chlorine residuals are difficult to maintain, particularly in Australia and the USA [1,2]. Monochloramine decay can occur due to both chemical and microbiological reactions. To manage disinfection residuals in drinking water, it is important to discriminate between chemical and microbiological decay processes [3,4]. The relative contribution of microbiological mediated monochloramine decay to the overall monochloramine decay can be determined via a microbial decay factor (F_m) described by Sathasivan, Fisher and Kastl [4]. Briefly, the microbial decay factor is determined from the ratio of the monochloramine decay rate of unfiltered water to filtered (0.2 μm membrane) water. In this method, the filtered water has microorganisms removed that are responsible for microbiological monochloramine decay, and represents decay by chemical means only. Conversely, in the unfiltered water, chloramine decay occurs by both microbial and chemical processes.

Using oxidants prior to chloramination for either transforming or eliminating chemical and microbial substances responsible for chloramine decay can be a viable strategy for water utilities to control the stability of monochloramine residual in water. Chlorine is the most commonly applied pre-oxidant in water treatment; however, due to the formation of regulated disinfection by-products (DBPs) from chlorine (i.e., trihalomethanes and haloacetic acids), the use of ultraviolet (UV) radiation, chlorine dioxide, and ozone has received attention within the last decade [5]. Ozone as a strong oxidant has been used in drinking water treatment for more than 100 years to control taste and odor, decrease precursors of DBPs, remove inorganic pollutants, and increase organic biodegradation [6,7]. However, ozone can be rapidly consumed under typical drinking water conditions. It cannot be used to maintain a disinfectant residual throughout the distribution system [8]. For this

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reason, most ozonation treatment scenarios include either chlorine or chloramine as a final disinfectant [9].

The motivations for inclusion of UV irradiation as a pre-oxidation process in chloraminated systems include the ability to inactivate most pathogenic microorganisms (particularly those resistant to chlorine such as Cryptosporidium) without forming DBPs at common disinfection doses. Therefore, a lower monochloramine dose can be used as a secondary disinfectant [10,11]. Although, it was reported that the effects of sequential disinfection with UV light and free chlorine or monochloramine (under practically relevant conditions) on DBPs formation varied among different NOM-containing waters, sequential exposure to UV and monochloramine were found to be the best option for dealing with UV resistant organisms [12,13]. In one study that evaluated UV-chloramine treatment, it was found that application of UV at disinfection doses had little effect on regulated DBPs. The combination of UV irradiation and chemical disinfectants such as chlorine or monochloramine has been shown to produce a synergistic disinfection effect and prevent subsequent microbial regrowth [10].

The effectiveness of pre-oxidants such as UV irradiation and ozone for controlling chemical and microbiological processes that have impact on decay of disinfectants in water has been an active area of research. Most previous studies with UV irradiation and ozone as pre-oxidants followed by another disinfectant (chlorine or chloramine) have focused on their potential in reducing or enhancing the DBPs formation [5,10,13–16]. However, the impacts of ozone and UV irradiation on different chloramine decay processes need more attention.

A map of specific fluorescence functionality of different organic substances in water samples can be achieved using a three dimensional fluorescence excitation emission matrix (3D-FEEM) spectroscopy. Excitation-emission matrices (EEM) can provide semi-quantitative characterisation information on dissolved organic matter (DOM) [17]. The 3D-FEEM spectroscopy measurement can also provide organic character information specifically on microbial protein-like and humic/fulvic-like substances in natural organic matter [17–19]. Previous studies on 3D-FEEM analysis have identified several common fluorescence peaks observed in water samples, attributed to proteins, humic and fulvic acids [20–23]. Strong correlations have been found between the fluorescence intensity of various peaks and water quality parameters such as total organic carbon (TOC), dissolved organic carbon (DOC), UV_{254}, ammonia (NH_3), and nitrate (NO_3^-) [24–27]. However, to the best of our knowledge, the relationship between the fluorescence intensity of various peaks and microbial decay factor (F_m) has not been investigated.

Considering the limited information available on the impacts of pre-oxidation processes on the microbiological processes responsible for monochloramine decay, the main objective of this study was to conduct a systematic investigation to assess the influence of UV irradiation and ozone as pre-oxidants on microbiologically assisted monochloramine decay. Ozone with varying contact times (0 to 50 mg min/L), and UV irradiation with doses up to 120 mJ/cm^2 were employed before chloramination for water samples with different water quality characteristics, and the monochloramine decay behavior was monitored to measure the microbial decay factor (F_m). In addition, efforts were made to identify and quantify the changes in different organic and microbial group compounds in water samples caused by selected pre-oxidation processes using 3D-FEEM spectroscopy, and correlations between the fluorescence peaks and F_m was achieved. To the best of our knowledge, the effects of UV light and ozone as pre-oxidation processes on microbiologically accelerated monochloramine decay has not previously studied.

2. Materials and methods

2.1. Water samples

Raw water samples were collected from the River Murray in South Australia (SA). In addition, conventionally treated water samples were collected prior to monochloramine disinfection, at the Tailem Bend water treatment plant (T.B WTP) in South Australia from 2015 to 2016. The T.B source water (from the River Murray) is treated by conventional treatment (coagulation/flocculation/sedimentation/filtration) followed by disinfection by UV irradiation and chloramination [2]. The UV disinfection system in T.B WTP is carried out by a continuous flow UV reactor using low pressure mercury lamps, currently targeting doses approximately 40 mJ/cm^2, as an additional barrier to *Cryptosporidium*. The efficacy of ozone contact periods and UV disinfection doses were evaluated individually using raw water from River Murray and treated water (before disinfection) from T.B WTP. In addition, water samples were also taken from before and after of the installed UV reactor in T.B WTP to investigate the impacts of the continuous flow UV reactor on microbiologically accelerated monochloramine decay. Pre-oxidated water samples (with a specific UV dose or ozone contact time) were then chloraminated in the laboratory and the rate of monochloramine decay over time was studied. All water samples were processed to determine the relative contribution of microbiological mediated monochloramine decay to the overall monochloramine decay in the bulk water, by determination of F_m described by Sathasivan, Fisher and Kastl [4].

2.2. Analytical methods

Water samples were ozonated in batch mode, and ozone demand and residual concentrations were measured using the Indigo colorimetric method where colour was measured at $\lambda = 600$ nm using a UV–vis Spectrophotometer (Shimadzu) [28]. Ozone stock solution was produced using the Ozone Generator BMT 803N. Potassium Indigo Trisulfonate, $C_{16}H_7N_2O_{11}S_3K_3$ was obtained from Sigma-Aldrich, USA. Ultrapure water was obtained from a Milli-Q purification system (Millipore, France). UV irradiation was performed using a UViFlo 4000 UV disinfection unit. Water samples were chloraminated under laboratory conditions using a free chlorine stock solution prepared by the addition of gaseous chlorine to ultra-pure water and analytical grade liquid ammonia (1000 mg/L as N). Chloramine residuals, pH, temperature and ammonia concentration for all water samples were analyzed at different times. Free chlorine and monochloramine were determined using N, N-diethyl-p-phenylenediamine (DPD) – ferrous ammonium sulphate (FAS) titrimetric procedure [29]. Ammonia concentrations were determined using ammonia-selective electrode Standard Method 4500-NH3 (D) [29]. UV absorbance at 254 nm (UVA_{254}) was measured through a 1 cm quartz cell using an Evolution 60 Spectrophotometer (Thermo Scientific, USA). Dissolved organic carbon (DOC) concentration was measured using a Sievers 900 Total Organic Carbon Analyser (GE Analytical Instruments, USA). Measurements of pH and temperature were conducted using a portable pH meter with a sealed, gel filled reference electrode with temperature compensation (pH320, WTW, Germany). 3D-FEEM spectra of water samples were acquired using a fluorescence spectrophotometer (PLS55, Perkin Elmer Instruments, Norwalk, USA). Emission (Em.) spectra were scanned from 280 to 540 nm at 0.5 nm increments and excitation (Ex.) spectra scanned from 250 to 400 nm with 5 nm increments [18]. The slits for excitation and emission were 5 nm. The Rayleigh scattering effect was minimized by subtracting the fluorescence spectra collected from a blank sample of deionized water.

2.3. Procedure for ozonation

An ozone stock solution was generated in a 1L ozone flask with 1 L of ultrapure water, and it was used to add ozone to the water samples. Aliquots of the water samples were accurately dosed with ozone-demand free water at a variety of concentrations. The water samples were pre-oxidised with ozone at different contact times (Ct's) up to 50 mg min/L. The kinetics of the initial decay of ozone was determined

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